

Temperature dependent dielectric relaxation measurement of an amino acid derivative based deep eutectic solvent: origin of timescales via experiment and molecular dynamics simulation

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Abstract

We report here temperature dependent ($303 \leq T/K \leq 343$) dielectric relaxation (DR) measurement of [Betaine+Urea+Water]¹ deep eutectic solvent (DES)² in the frequency window from 20Hz to 50 GHz (with a gap from 10 MHz to 200 MHz). We also performed molecular dynamics simulation of DR in the above-mentioned temperature range employing General Amber Force Field (GAFF)³ model interaction. Experimental DR measurement of this DES require multi-debye fits and produce well separated DR time scales that are spread over sub-10 picosecond to few nanoseconds (~ 3 ns). A comparison between simulation predictions and the experimental DR data in the KHz-GHz frequency regime describes the well similarities of the dynamics(time-scales). A well correlation between the measured DR and simulated DR activation energies further reveals the strong connection between the measured DR and the simulated $C_M(t)$ (dipole moment-moment autocorrelation function). The simulated single dipole reorientation dynamics also contributes to the collective polarization relaxation. Average experimental DR times exhibits fractional viscosity dependence. Deviation from hydrodynamic predictions confirms partial decoupling of the solute rotation from the medium frictional resistance exerted by the viscosity. Also, viscosity related activation energy ($E_a=57$ kJ/mol) is ~ 1.5 times larger than activation energy related to experimental average DR times ($E_a=39$ kJ/mol) which supports the fractional viscosity dependence of the medium dynamics.

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